

# Effects of absorption on multiple scattering by random particulate media: exact results

Michael I. Mishchenko<sup>1\*</sup>, Li Liu<sup>1</sup>, and Joop W. Hovenier<sup>2</sup>

<sup>1</sup>NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025

<sup>2</sup>Astronomical Institute "Anton Pannekoek", University of Amsterdam, Kruislaan 403, 1098 SJ Amsterdam, The Netherlands

\*[mmishchenko@giss.nasa.gov](mailto:mmishchenko@giss.nasa.gov)

**Abstract:** We employ the numerically exact superposition  $T$ -matrix method to perform extensive computations of electromagnetic scattering by a volume of discrete random medium densely filled with increasingly absorbing as well as non-absorbing particles. Our numerical data demonstrate that increasing absorption diminishes and nearly extinguishes certain optical effects such as depolarization and coherent backscattering and increases the angular width of coherent backscattering patterns. This result corroborates the multiple-scattering origin of such effects and further demonstrates the heuristic value of the concept of multiple scattering even in application to densely packed particulate media.

©2007 Optical Society of America

**OCIS codes:** (030.1670) Coherent optical effects; (030.5620) Radiative transfer; (290.4210) Multiple scattering; (290.5850) Scattering, particles.

---

## References and links

1. S. H. Tseng, A. Taflov, D. Maitland, and V. Backman, "Pseudospectral time domain simulations of multiple light scattering in three-dimensional macroscopic random media," *Radio Sci.* **41**, RS4009 (2006).
2. D. W. Mackowski, "Direct simulation of scattering and absorption by particle deposits," *Proc. IMECE* **2006**, 14615 (2006).
3. M. I. Mishchenko and L. Liu, "Weak localization of electromagnetic waves by densely packed many-particle groups: exact 3D results," *J. Quant. Spectrosc. Radiat. Transfer* **106**, 616–621 (2007).
4. M. I. Mishchenko, L. Liu, D. W. Mackowski, B. Cairns, and G. Videen, "Multiple scattering by random particulate media: exact 3D results," *Opt. Express* **15**, 2822–2836 (2007).
5. A. Penttilä and K. Lumme, "Coherent backscattering effects with Discrete Dipole Approximation method," in *Peer-Reviewed Abstracts of the Tenth Conference on Electromagnetic & Light Scattering*, G. Videen, M. Mishchenko, M. P. Mengüç, and N. Zakharova, eds. (<http://www.giss.nasa.gov/~crimim/>, 2007), pp. 157–160.
6. J. W. Hovenier, C. van der Mee, and H. Domke, *Transfer of Polarized Light in Planetary Atmospheres – Basic Concepts and Practical Methods* (Springer, Berlin, 2004).
7. M. I. Mishchenko, L. D. Travis, and A. A. Lacis, *Multiple Scattering of Light by Particles: Radiative Transfer and Coherent Backscattering* (Cambridge U. Press, Cambridge, UK, 2006).
8. H. C. van de Hulst, *Multiple Light Scattering. Tables, Formulas, and Applications* (Academic Press, San Diego, 1980).
9. S. Etemad, R. Thompson, M. J. Andrejco, S. John, and F. C. MacKintosh, "Weak localization of photons: termination of coherent random walks by absorption and confined geometry," *Phys. Rev. Lett.* **59**, 1420–1423 (1987).
10. M. I. Mishchenko, L. D. Travis, and A. A. Lacis, *Scattering, Absorption, and Emission of Light by Small Particles* (Cambridge U. Press, Cambridge, UK, 2002). <http://www.giss.nasa.gov/~crimim/books.html>.
11. F. M. Kahnert, "Numerical methods in electromagnetic scattering theory," *J. Quant. Spectrosc. Radiat. Transfer* **79–80**, 775–824 (2003).
12. D. W. Mackowski and M. I. Mishchenko, "Calculation of the  $T$  matrix and the scattering matrix for ensembles of spheres," *J. Opt. Soc. Am. A* **13**, 2266–2278 (1996).
13. D. Mackowski, K. Fuller, and M. Mishchenko, "Codes for calculation of scattering by clusters of spheres." <ftp://ftp.eng.auburn.edu/pub/dmckowski/scatcodes/index.html>.
14. Y. Kuga and A. Ishimaru, "Retroreflectance from a dense distribution of spherical particles," *J. Opt. Soc. Am. A* **1**, 831–835 (1984).
15. L. Tsang and A. Ishimaru, "Backscattering enhancement of random discrete scatterers," *J. Opt. Soc. Am. A* **1**, 836–839 (1984).

## 1. Introduction

Several recent publications [1–5] have marked the emergence of an accurate quantitative approach to the problem of light scattering by macroscopic media composed of randomly positioned particles based on numerically exact solutions of the Maxwell equations. In particular, in [4] we simulated the onset and evolution of various multiple-scattering effects by increasing the number of particles,  $N$ , in a statistically homogeneous volume of discrete random medium. We argued that if specific scattering features either intensify or weaken with  $N$  in an expected way [6, 7] then these features can be attributed to the increasing effect of multiple scattering. The particles in [4] were assumed to be non-absorbing by assigning to them a real-valued relative refractive index. However, it is well known that absorption can be an efficient suppressant of multiple scattering [6–8] and reduces the average length of multiple-scattering paths [9]. It is, therefore, important to verify whether the multiple-scattering interpretation of the various optical displays in [4] is consistent with the way in which these features should be expected to change upon increased absorption. This verification is the main objective of the present paper.

## 2. Concept of multiple scattering

Before describing and discussing the results of numerical computations, it is instructive to explain the primordial origin of the concept of multiple scattering. The majority of theoretical techniques based on a direct solution of the frequency-domain differential Maxwell equations or their integral counterparts are applicable to an arbitrary fixed finite object, be it a single physical body or a cluster consisting of several distinct components [10, 11]. These techniques are based on treating the object as a single scatterer and yield the cumulative scattered electric field. However, if the object is a multi-particle group, such as a cloud of water droplets, then it is often convenient to represent the scattered field at a point  $\mathbf{r}$  as a superposition of partial fields scattered by the individual particles, i.e.,

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \sum_{i=1}^N \mathbf{E}_i^{\text{sca}}(\mathbf{r}), \quad (1)$$

where  $N$  is the number of particles in the group,  $\mathbf{E}(\mathbf{r})$  is the total field,  $\mathbf{E}^{\text{inc}}(\mathbf{r})$  is the incident field, and  $\mathbf{E}_i^{\text{sca}}(\mathbf{r})$  is the  $i$ th partial scattered field. The partial scattered fields can be found by solving the vector Foldy–Lax equations (FLEs) [7]. Specifically, the  $i$ th partial scattered field is given by

$$\mathbf{E}_i^{\text{sca}}(\mathbf{r}) = \int_{V_i} d\mathbf{r}' \tilde{G}(\mathbf{r}, \mathbf{r}') \cdot \int_{V_i} d\mathbf{r}'' \tilde{T}_i(\mathbf{r}', \mathbf{r}'') \cdot \mathbf{E}_i(\mathbf{r}''), \quad (2)$$

where  $V_i$  is the volume occupied by the  $i$ th particle,  $\tilde{G}(\mathbf{r}, \mathbf{r}')$  is the free-space dyadic Green's function, and  $\mathbf{E}_i(\mathbf{r}'')$  is the electric field "exciting" particle  $i$ . The  $N$  dyadics  $\tilde{T}_i$  can be found by solving the following equation for each  $i$  separately:

$$\tilde{T}_i(\mathbf{r}, \mathbf{r}') = k_1^2 [m_i^2(\mathbf{r}) - 1] \delta(\mathbf{r} - \mathbf{r}') \tilde{I} + k_1^2 [m_i^2(\mathbf{r}) - 1] \int_{V_i} d\mathbf{r}'' \tilde{G}(\mathbf{r}, \mathbf{r}'') \cdot \tilde{T}_i(\mathbf{r}'', \mathbf{r}'), \quad \mathbf{r}, \mathbf{r}' \in V_i, \quad (3)$$

where  $k_1$  is the wave number in the host medium,  $m_i(\mathbf{r})$  is the relative refractive index, and  $\tilde{I}$  is the identity dyadic. The  $\tilde{T}_i$  is the dyadic transition operator of particle  $i$  with respect to the fixed laboratory coordinate system computed in the absence of all the other particles. In other words, the  $N$  dyadic transition operators are totally independent of each other. However,

the exciting fields are interdependent and must be found by solving the following system of  $N$  linear integral equations:

$$\mathbf{E}_i(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \sum_{j(\neq i)=1}^N \int_{V_j} d\mathbf{r}' \tilde{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \int_{V_j} d\mathbf{r}'' \tilde{\mathbf{T}}_j(\mathbf{r}', \mathbf{r}'') \cdot \mathbf{E}_j(\mathbf{r}''), \quad \mathbf{r} \in V_i, \quad i=1, \dots, N. \quad (4)$$

Let us rewrite Eqs. (1), (2), and (4) in the following compact operator form:

$$E = E^{\text{inc}} + \sum_{i=1}^N \hat{G}\hat{T}_i E_i, \quad (5)$$

$$E_i = E^{\text{inc}} + \sum_{j(\neq i)=1}^N \hat{G}\hat{T}_j E_j, \quad (6)$$

where

$$\hat{G}\hat{T}_j E_j = \int_{V_j} d\mathbf{r}' \tilde{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \int_{V_j} d\mathbf{r}'' \tilde{\mathbf{T}}_j(\mathbf{r}', \mathbf{r}'') \cdot \mathbf{E}_j(\mathbf{r}''). \quad (7)$$

Iterating Eq. (6) yields

$$E_i = E^{\text{inc}} + \sum_{j(\neq i)=1}^N \hat{G}\hat{T}_j E^{\text{inc}} + \sum_{\substack{j(\neq i)=1 \\ l(\neq j)=1}}^N \hat{G}\hat{T}_j \hat{G}\hat{T}_l E^{\text{inc}} + \sum_{\substack{j(\neq i)=1 \\ l(\neq j)=1 \\ m(\neq l)=1}}^N \hat{G}\hat{T}_j \hat{G}\hat{T}_l \hat{G}\hat{T}_m E^{\text{inc}} + \dots, \quad (8)$$

while the substitution of Eq. (8) in Eq. (5) gives what can be called an order-of-scattering expansion of the total electric field:

$$E = E^{\text{inc}} + \sum_{i=1}^N \hat{G}\hat{T}_i E^{\text{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1}}^N \hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\text{inc}} + \sum_{\substack{i=1 \\ j(\neq i)=1 \\ l(\neq j)=1}}^N \hat{G}\hat{T}_i \hat{G}\hat{T}_j \hat{G}\hat{T}_l E^{\text{inc}} + \dots. \quad (9)$$

Indeed,  $\hat{G}\hat{T}_i E^{\text{inc}}$  can be interpreted as the partial scattered field at the observation point generated by particle  $i$  in response to the excitation by the incident field only,  $\hat{G}\hat{T}_i \hat{G}\hat{T}_j E^{\text{inc}}$  is the partial field generated by the same particle in response to the excitation caused by particle  $j$  in response to the excitation by the incident field, etc. This order-of-scattering interpretation of Eq. (9) becomes even more transparent when the particles are widely separated, and single-scattering dyadics replace the dyadic transition operators as complete electromagnetic descriptors of the individual particles [7].

It is important to recognize that besides being an interpretation and visualization tool, the concept of multiple scattering does not represent a physical process per se in the framework of frequency-domain electromagnetics. Indeed, it follows from Eq. (4) that all mutual particle-particle excitations occur simultaneously and are not temporally discrete and ordered events. Nevertheless, Eq. (9) constitutes a very fruitful way of re-writing the original FLEs, while the “multiple scattering” terminology is a convenient and compact way of illustrating and interpreting their solutions [4].

### 3. Numerical results

It is well known from the theory of radiative transfer in homogeneous plane-parallel atmospheres [6–8] that the contribution of light scattered  $n$  times to the total specific intensity is proportional to the  $n$ th power of the single-scattering albedo. The single-scattering albedo is equal to unity for  $\text{Im}(m) = 0$  but can decrease significantly as  $\text{Im}(m)$  increases, thereby

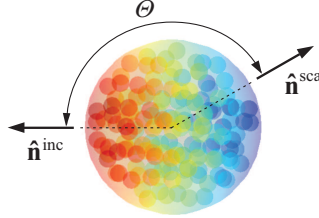


Fig. 1. Far-field electromagnetic scattering by a volume of discrete random medium.

suppressing various manifestations of multiple scattering. The radiative transfer theory and the concept of the single-scattering albedo may not be applicable directly to densely packed particles. Still, the various effects identified in [4] as being caused by multiple scattering should be expected to diminish and ultimately vanish with increasing absorption. Indeed, if absorption diminishes the response of a particle to a certain excitation, this effect should have a stronger influence on multiply scattered radiation than on singly scattered radiation: the  $n$ -th order of scattering would suffer  $n$  times in succession from such a diminished response, Eq. (9).

To verify whether this is indeed the case, we have performed numerically exact computations of far-field scattering by a spherical volume of discrete random medium filled with 160 identical spherical particles (Fig. 1). The size parameters of the volume and the particles are fixed at  $k_1 R = 40$  and  $k_1 r = 4$ , respectively, which means that the particle volume concentration is equal to 16%. The real part of the particle refractive index is fixed at 1.32, while the imaginary part is varied from 0 to 0.3. To perform averaging over particle positions, we use only one randomly configured 160-particle group and average over all possible orientations of this configuration with respect to the laboratory coordinate system [4]. This approach yields an infinite continuous set of random realizations of the 160-particle group and allows us to employ the highly efficient orientation averaging technique afforded by the superposition  $T$ -matrix method [10, 12, 13].

We assume that the statistically random particulate volume is illuminated by a plane electromagnetic wave or a parallel quasi-monochromatic beam of light propagating in the direction  $\hat{n}^{\text{inc}}$  (Fig. 1). The observation direction is specified by the unit vector  $\hat{n}^{\text{sca}}$ . Since all scattering properties of the volume are averaged over the uniform orientation distribution of the multi-particle group, we can simplify the discussion by using the scattering plane for defining the Stokes parameters of the incident and scattered light. The transformation of the Stokes parameters in the far-field zone of the volume is then written in terms of the normalized Stokes scattering matrix [4, 6, 7]:

$$\begin{bmatrix} I^{\text{sca}} \\ Q^{\text{sca}} \\ U^{\text{sca}} \\ V^{\text{sca}} \end{bmatrix} \propto \begin{bmatrix} a_1(\theta) & b_1(\theta) & 0 & 0 \\ b_1(\theta) & a_2(\theta) & 0 & 0 \\ 0 & 0 & a_3(\theta) & b_2(\theta) \\ 0 & 0 & -b_2(\theta) & a_4(\theta) \end{bmatrix} \begin{bmatrix} I^{\text{inc}} \\ Q^{\text{inc}} \\ U^{\text{inc}} \\ V^{\text{inc}} \end{bmatrix}, \quad (10)$$

where  $\theta$  is the scattering angle (Fig. 1). The phase function is normalized according to

$$\frac{1}{2} \int_0^\pi d\theta \sin \theta a_1(\theta) = 1. \quad (11)$$

The most relevant numerical results are shown in Fig. 2. For comparison, the yellow curves depict the results computed for a single sphere with  $k_1 r = 4$  and  $m = 1.32 + i0.3$ .

Note that increasing  $\text{Im}(m)$  above a certain threshold can cause decreasing rather than increasing absorption since the scatterer eventually behaves as a metallic object. Therefore,

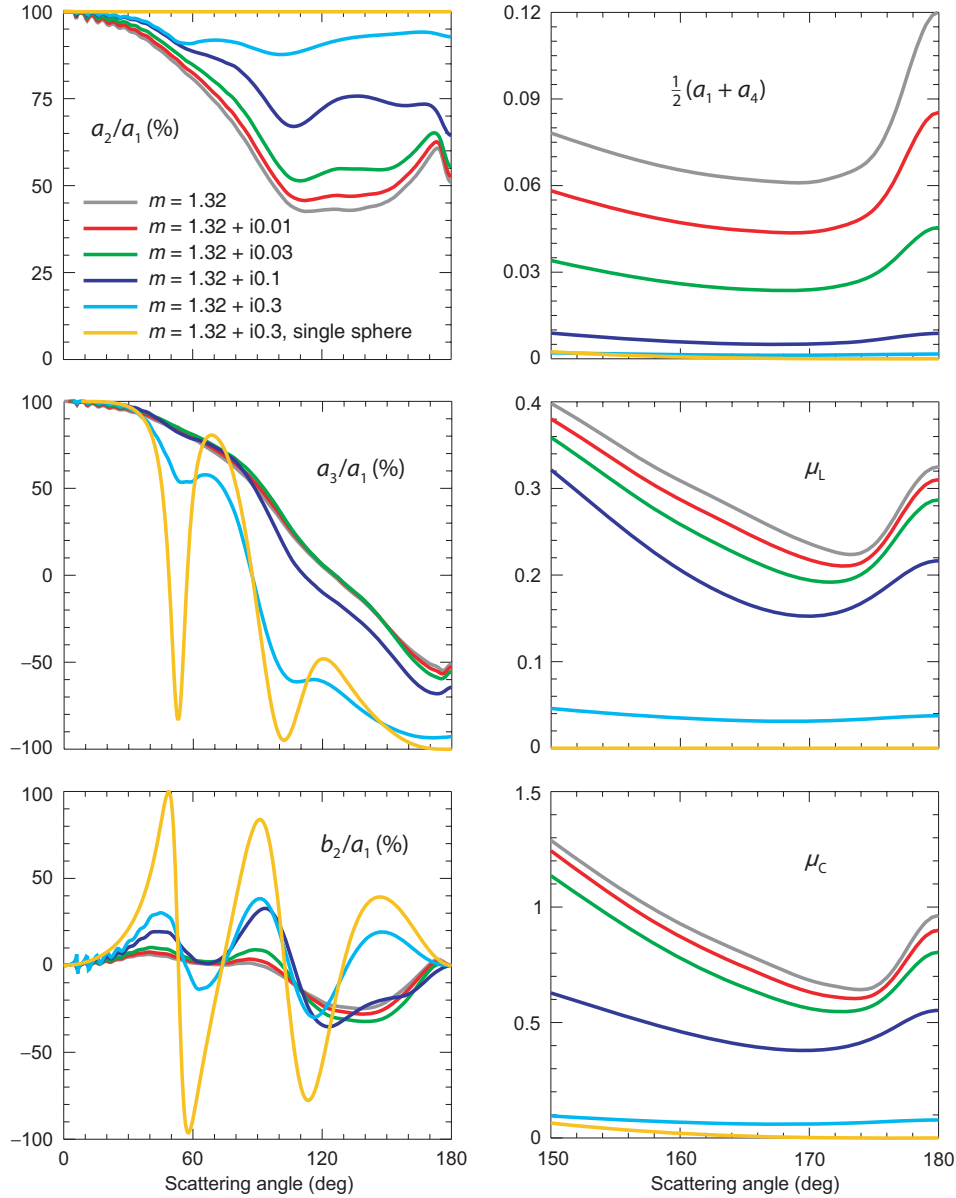


Fig. 2. Far-field scattering properties of a volume of discrete random medium.

we have verified specifically that the single-scattering albedo of the entire scattering volume decreases and the total absorption cross section increases as  $\text{Im}(m)$  is increased from 0 to 0.3.

#### 4. Discussion

The ratio  $a_2(\Theta)/a_1(\Theta)$  is identically equal to unity for scattering by a single sphere. Therefore, the large deviation of this ratio from 100% for a scattering volume comprising non-absorbing particles was interpreted in [4] as being a direct consequence of the strengthened depolarizing effect of multiple scattering. The upper left panel of Fig. 2 obviously reinforces this interpretation. Indeed, the deviation of  $a_2(\Theta)/a_1(\Theta)$  from 100% decreases with

increasing  $\text{Im}(m)$  quite significantly, even though it does not completely vanish even for  $\text{Im}(m) = 0.3$  thereby revealing a residual influence of multiple scattering. The deviation of the ratio  $a_3(180^\circ)/a_1(180^\circ)$  from  $-100\%$  behaves quite similarly. With increasing absorption, the ratios  $a_2(\theta)/a_1(\theta)$ ,  $a_3(\theta)/a_1(\theta)$ , and  $b_2(\theta)/a_1(\theta)$  for the entire scattering volume should be expected to approach those for a single constituent sphere. The left-hand panels of Fig. 2 clearly exhibit this tendency, although the residual multiple-scattering effect diminishes noticeably the large amplitude of the interference features typical of monodisperse spheres. The increasing amplitude of oscillations of the ratio  $b_2(\theta)/a_1(\theta)$  with increasing  $\text{Im}(m)$  and the similar behavior of the ratio  $-b_1(\theta)/a_1(\theta)$  (not shown) obviously represent what is known in astrophysics of planetary surfaces as the Umov effect.

An important, inherently multiple-scattering effect is coherent backscattering (CB) [4, 7, 14–16]. The grey curves in the right-hand panels of Fig. 2 demonstrate the manifestations of CB in the same-helicity scattered intensity

$$\frac{1}{2}(I^{\text{sca}} + V^{\text{sca}}) \propto \frac{1}{2}[a_1(\theta) + a_4(\theta)] \quad (12)$$

as well as in the linear and circular polarization ratios defined, respectively, by

$$\mu_L = \frac{I^{\text{sca}} - Q^{\text{sca}}}{I^{\text{sca}} + Q^{\text{sca}}} = \frac{a_1(\theta) - a_2(\theta)}{a_1(\theta) + 2b_1(\theta) + a_2(\theta)} \quad (13)$$

and

$$\mu_C = \frac{I^{\text{sca}} + V^{\text{sca}}}{I^{\text{sca}} - V^{\text{sca}}} = \frac{a_1(\theta) + a_4(\theta)}{a_1(\theta) - a_4(\theta)}. \quad (14)$$

It is clearly seen that increasing absorption diminishes and then essentially extinguishes these features, which corroborates their multiple-scattering origin.

Further evidence comes from the examination of the dependence of the angular width of CB patterns on absorption. It is well known that absorption terminates long scattering paths and thereby can be expected to reduce the range of angles affected by CB [9]. This is clearly seen indeed, especially in the panels depicting  $\mu_L$  and  $\mu_C$ .

## 5. Conclusion

The concept of multiple scattering is native to the theories of radiative transfer and CB describing electromagnetic energy transport in low-density particulate media. In fact, the very derivation of the radiative transfer equation from the Maxwell equations is explicitly based on the order-of-scattering expansion of the far-field FLEs [7]. One might argue that the relevance of this concept to densely packed particulate media is not obvious and may not be easy to justify. Our previous results [3, 4] have demonstrated that the evolution of certain scattering patterns with increasing the number of particles in a volume of discrete random medium is qualitatively consistent with the expected result of intensifying multiple scattering. Our new numerical data show that increasing absorption diminishes and nearly extinguishes these scattering patterns and increases the angular width of the CB patterns, thereby further corroborating their multiple-scattering origin. This result provides an additional illustration of the heuristic value of the concept of multiple scattering, even when this concept is applied to densely packed particulate media.

## Acknowledgments

The authors thank Daniel Mackowski, Michiel Min, and Gordon Videen for numerous useful discussions. This research was supported by the NASA Radiation Sciences Program managed by Hal Maring and by the NASA Glory Mission project.